To: Craig Beck

Paper

PLUTONIUM CONTAMINATION IN SOILS IN OPEN SPACE AND RESIDENTIAL AREAS NEAR ROCKY FLATS, COLORADO

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Abstract—Spatial analysis of the ^{240}Pu:^{239}Pu isotopic ratio of 42 soil samples collected around Rocky Flats Plant near Golden, Colorado, was conducted to assess the effect of Rocky Flats Plant activity on the soil environment. Two probability maps that quantified the uncertainty of the spatial distribution of plutonium isotopic ratios were constructed using the sequential Gaussian simulation technique (sGs). Assuming a plutonium isotopic ratio range of 0.152±0.003 to 0.169±0.009 is characteristic to global fallout in Colorado, and a mean value of 0.155 is representative for the Rocky Flats Plant area, the main findings of the current work were (1) the areas northwest and southwest of Rocky Flats Plant exhibited a plutonium ratio ≥ 0.155, thus were minimally impacted by the plant activity; (2) the study area east of Rocky Flats Plant (~ 120 km^2) exhibited a plutonium isotopic ratio ≤ 0.155, which is a definitive indicator of Rocky Flats Plant-derived plutonium; and (3) inventory calculations across the study area exhibited large standard error of estimates. These errors were originated from the high variability in plutonium activity over a small sampling scale and the uncertainty in the global fallout isotopic ratio. Using the mean simulated estimates of plutonium isotopic ratio, coupled with plutonium activity measured at 11 soil pits and additional plutonium information published elsewhere, the plutonium loading on the open space and residential areas amounted to 111.2 GBq, with a standard error of estimate of 50.8 GBq.

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Key words: soil; plutonium; contamination, environmental; sampling

INTRODUCTION

Plutonium contamination of soils around Rocky Flats Plant (RFP), Colorado, resulted mainly from past outdoor-storage practices at a site locally known as the 903 Pad and subsequent remobilization due to inadequate cleanup methods (Krey and Hardy 1970; Seed et al. 1971; Little et al. 1980). Until now, cleanup decisions and litigation issues regarding off-plant areas have not been resolved because of a lack of sufficient information regarding the nature and extent of $^{239+240}Pu$ and ^{241}Am in soils.

The Remedial Investigations (RI) of the soil environment outside RFP initiated in 1992 and included characterization of the spatial distribution of $^{239+240}Pu$ (Litaor et al. 1995) and of ^{241}Am (Litaor and Allen 1996). The spatial analysis of plutonium was performed using four different data sets, one of which contained 852 samples collected between 1974 and 1994. This data set is the most comprehensive compilation of plutonium data with unique coordinates from the RFP vicinity ever assembled. The technique of nonparametric indicator kriging was used to model the conditional cumulative distribution function (ccdf) of plutonium in soils around RFP. The ccdf was used to generate an E-type (mean of the conditional cdf) surface and a map of the probability of exceedance of plutonium global fallout around RFP. The resulting surfaces were consistent with the hypothesis that the westerly winds are the dominant mechanism of plutonium dispersal. Other processes, such as downstream transport of sediment along local southeast-trending drainages, may have additionally moved small amounts of plutonium.

Although the above work is geostatistically robust, the critical issue of the relative contribution of RFP-derived plutonium vs. global fallout in open space and residential areas around RFP is still pending. This information is also required by the Dose Reconstruction Study conducted by Colorado Department of Public Health and Environment (CDPHE) and for the litigation proceedings currently in progress.

Krey (1976) conducted a plutonium isotopic ratio study around RFP that paid much attention to the reproducibility and reliability of plutonium analysis and the precision of the mass spectrometric analysis. Only scant attention was given to the spatial variability of the plutonium isotopic ratio as well as reproducibility of the plutonium analysis in various sampling scales. Krey (1976) suggested that a plutonium isotopic ratio ≥ 0.163 represents a global-fallout plutonium. As part of the geochemical characterization study conducted by DOE (U.S. DOE 1995), twelve soils were collected from locations believed to represent global fallout in Colorado. These soils were analyzed and exhibited a ^{240}Pu:^{239}Pu ratio of 0.155 ± 0.019. Efurd et al. (1995) analyzed 35 soil samples that represent a larger subset of the background soils collected along the Colorado Front Range

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..nd reported a value of 0.169 ± 0.005 for the global fallout in Colorado. Ibrahim et al. (1997) collected two soils believed to represent global fallout and found a ^{240}Pu:^{239}Pu isotopic ratio of 0.152 ± 0.003. This survey clearly shows that there is no unique value that best represents global fallout in Colorado, thus a range of values between 0.152 to 0.169 can be used as global fallout. In the present study, the value of 0.155 reported by DOE (U.S. DOE 1995) was selected to represent global fallout around RFP. If plutonium ratio of 0.155 represents the global fallout and ^{240}Pu:^{239}Pu ratio of 0.051 ± 0.009 denotes a weapon-grade plutonium processed in Rocky Flats (Krey and Krajewski 1972) any ^{240}Pu:^{239}Pu ratio between these two values represents some mixture of global fallout and Rocky Flats plutonium. A new study of the plutonium isotopic ratio was initiated in 1994, to produce probability maps of plutonium isotopic ratio in open space and residential areas potentially impacted by RFP activity, and to compute $^{239+240}Pu$ loading on these areas.

Field and laboratory methods

The optimal number of soil samples and the optimal distance between the plots in the open space and residential areas were determined using a sampling strategy algorithm for soil sampling suggested by McBratney et al. (1981). The computed optimum sampling interval for ..e grid spacing between plots within the open space and ..esidential areas near RFP was 1,750 m. To reduce the estimation variance even further, the optimal sampling interval was decreased to 1,000 m between grid plots. The actual location of the plots was impacted by past and present land-use practices, access refusal by land owners, and significant growth in residential and commercial areas east of RFP during the last 20 y.

Sixty-three samples were collected using the CDH and RFP sampling techniques (Litaor et al. 1995) and shipped to Los Alamos National Laboratory for ^{240}Pu:^{239}Pu ratio determination. Thermal ionization mass spectrometry (TIMS) was used to measure the atoms of ^{239}Pu and ^{240}Pu separately. A detailed account of the analytical technique and the pertinent quality assurance and quality control (QA/QC) information is given by Efurd et al. (1995).

For an analysis of the plutonium inventory, 11 soils east of RFP were excavated, described, and sampled. The soil pits were excavated at undisturbed or minimally disturbed sites determined by vegetation composition and association. Sampling soil for plutonium characterization involves several special considerations: (1) potential cross-contamination of subsurface horizons from the more contaminated surface horizons; (2) collection of sufficient material to obtain representative plutonium activity and other soil parameters; and (3) selection of a realistic sampling design that considers the high cost of ..lutonium analysis and provides sufficient information ..egarding the vertical distribution of plutonium in the soil profile.

In light of these considerations, a special sampling method was employed, which involved digging a pit, 3 m to 5 m long, 1 m wide, and 1 m to 1.1 m deep. The vegetation at the surface of the pit wall selected for sampling was clipped close to the ground and discarded. The surface of the selected wall was then thoroughly scraped with a stainless steel spade to reduce the possibility of cross-contamination. Ten soil samples were collected per pit, according to the following depth intervals (in cm): 0–3, 3–6, 6–9, 9–12, 12–18, 18–24, 24–36, 36–48, 48–72, and 72–96. A bottom-to-top sampling sequence was adopted to reduce further the risk of cross contamination. Each soil sample was collected from within a horizontal cavity dug into the pit face at a selected depth. An exception to the above sequence was made for near-surface samples (0–12 cm) where the soil was too friable to permit discrete sampling. To sample the top section of the profile, the sampling was begun at ground level using a knife and spatula to cut an area approximately 25 cm long, 20 cm wide, and 3 cm deep. The entire soil mass in this area including roots and partially decomposed organic material was collected. Sampling continued in this manner for intervals as deep as 12 cm.

Sampling for selected physical and chemical parameters was conducted by genetic horizons rather than by the incremental depth procedure. The soil bulk density was measured using the clod method described by Blake and Hartge (1986). The clod method required a soil aggregate stable enough to cohere coating, weighing, and handling. It usually does not include rocks larger than 1–2 cm. The soils were described according to guidelines established by the Soil Survey Staff (1975, 1993) and classified as Aridic Argiustolls (TR 27, 31), Typic Argiaquolls (TR 28), Pachic Argiustolls (TR 32, 33, 37), Aridic Haplustolls (TR 29), Fluvaquentic Endoaquolls (TR 30), Torrertic Argiustolls (TR 34), Torrifluventic Haplustolls (TR 35), and Pachic Calciustolls (TR 36). For further details regarding the soil taxonomy and classification, see U.S. DOE (1996).

Geostatistical analysis

The spatial analysis of the plutonium isotopic ratio (^{240}Pu:^{239}Pu) was conducted using the stochastic simulation methodology described by Deutsch and Journel (1992). Simulation differs from ordinary kriging (Litaor 1995) and indicator kriging (Litaor et al. 1995) in two fundamental respects: (1) Kriging algorithm provides a "best"-local estimate of $Pu'(u)$ of each unsampled value at location (u) taken one at the time without specific attention to the resulting spatial statistics. Kriging estimates based on sparse soil samples may produce a strong smoothing effect especially in the areas of sparse sampling. In simulation, the resulting global features and statistics of the simulated values are the major goal of the analysis and the smoothing effect in areas of sparse sampling is greatly reduced (Deutsch and Journal 1992); (2) For a given conditional cumulative distribution function (ccdf) of plutonium isotopic ratio over a given area, the use of kriging as an interpolation algorithm produces a single numerical model. This model is "best" for the

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purpose of local accuracy. Simulation provides many alternative models (L), each of which is a "good" representation of the reality in some global sense. The differences among these L alternative models, also known as realizations, provide a measure of joint spatial uncertainty. The quantification of this uncertainty is the main motive for using stochastic simulation in assessing the spatial structure of the plutonium isotopic ratio across the open space and residential areas adjacent to RFP.

The most straightforward algorithm for generating realizations of the ^{240}Pu:^{239}Pu ratio is a sequential Gaussian simulation (Fig. 1). In brief, the plutonium isotopic ratio was simulated sequentially following its ccdf using a simple kriging (SK) system of equations.

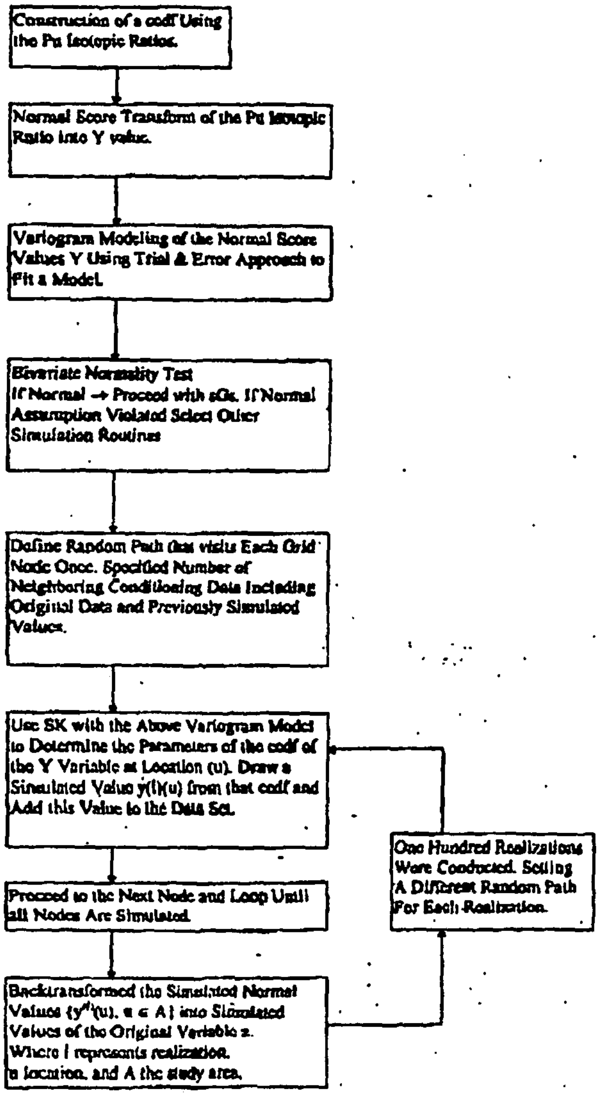

Fig. 1. Flow chart diagram of the sequential Gaussian simulation (sGs).

The conditioning data consist of all original data and all previously simulated values found within a neighborhood of the location being simulated. Prior to the simulation the data set was transformed using a normal score transform routine, and a bivariate normality check was performed. The normal score transform is a function that transforms the non-normal measured data into standard Gaussian cdf with zero mean and unit variance. The normal score transform defines a new variable Y, which is by construction univariate and normally distributed. Because the sGs assumes multivariate normal distribution across a study site, the bivariate normality test checks the bivariate cdf of any pair of values $Y(u)$, $Y(u+h)$ across the study site. The u designates georeferenced location, and h is the lag (distance) between two measurements. In practice, if the bivariate distribution test is normal, one can assume that multivariate Gaussian distribution of the random variable (e.g., plutonium isotopic ratio across the study site) will hold. The spatial analysis of the Y variable was conducted using a variogram routine and a trial and error approach to fit a model to the experimental variogram. The trial and error exercise is the preferred practice by most geostatisticians because it requires a prior understanding of the spatial arrangement of the data and a wise selection of the variogram parameters such as the range and nugget effect.

There are several steps involved in the execution of the sGs. First, a random path that visited each node at a selected grid was defined. The final grid, which covered the area east of State Plane Coordinate Projection 2093000, consisted of 10,000 nodes with node's size of 140 × 140 m. At each node a specified number of neighboring conditioning data included both original data and previously simulated grid node values. The final specifications were set to 4 for a minimum and 8 for maximum number of original data and 16 for previously simulated data. This specification represents an optimum number of nodes that were based on the variogram range and the spatial arrangement of the original data.

The next step included the use of a SK system of equations coupled with the normal score variogram model to compute the mean and the variance of the ccdf of the ^{240}Pu:^{239}Pu ratio at location u. A simulated value $Pu^{(A)}(u)$ was generated from that ccdf, and that simulated value was added to the data set. The next node was visited and looped until all nodes were simulated. This sequence of steps produced the first realization. To built multiple realizations, the previous sequence was repeated 100 times with a different random path for each realization. The simulated normal values of the 100 sequential Gaussian simulations were backtransformed for the original variable. During the backtransformed calculations within-class interpolations and tail extrapolations were called for. In the current analysis the hyperbolic model for upper tail model was used because of the positively skewed ccdf, and the linear model was used for the lower tail. The post-processing analysis can produce three output files. The first is an E-type estimate, which is a

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point-by-point average of the multiple realizations. Because the E-type estimates are average values, the 100 simulations is considered a reasonable number of realizations. The second output file contains a probability of exceeding a fixed cutoff value (e.g., background isotopic ratio), and the third output file consists of computed values when a fixed ccdf value p is reached (i.e., the conditional p-quantile value).

The overall loading (inventory) of plutonium in the open space and residential areas was computed using the E-type map derived from the sGs analysis coupled with plutonium activity and bulk density measurements performed on the 11 soil pits described above. Additional information regarding plutonium distribution with depth was gleaned from 14 macroplots studied by Webb et al. (1994). Because of the variable sampling scheme employed by Webb et al. (1994), only the pertinent data that fit the sampling sequence of the 11 soil pits were used. Exception was made for the areas bracketed by isopleth 0.10 and greater (see below), where a sampling depth of 0–21 cm was employed.

RESULTS AND DISCUSSION

Spatial analysis

The spatial distribution and statistics of the plutonium isotopic ratio from 42 soil samples are depicted in Figs. 2 and 3 and Table 1. The sites denoted by the dark cross exhibited a plutonium isotopic ratio range of 0.055 to 0.079 and represent the area most impacted by RFP activity. This area is adjacent to the plant perimeter and for the most part is restricted to the east and southeast directions. The sites denoted by a clear diamond exhibited a plutonium isotopic ratio range of 0.08 to 0.154,

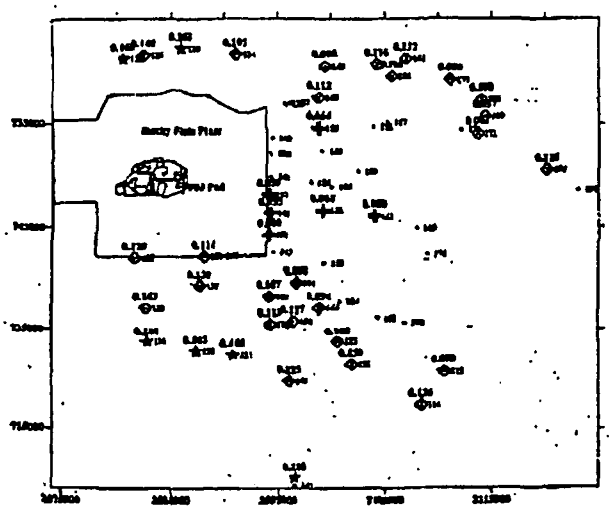

Fig. 2. Location and magnitude of ^{240}Pu:^{239}Pu isotopic ratios collected from 42 soil locations adjacent to RFP. ID numbers without isotopic ratio represent soil locations that were sampled but were not analyzed.

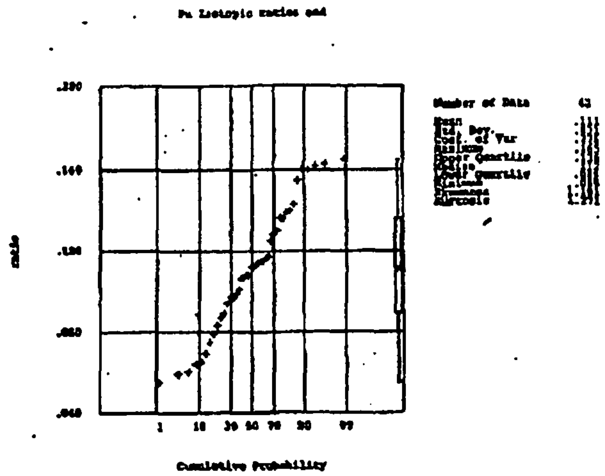

Fig. 3. The conditional cumulative distribution function (ccdf) of the plutonium isotopic ratio.

which represents a mixture of RFP plutonium with global fallout. The sites denoted by a dark star exhibited a plutonium isotopic ratio equal to or greater than 0.155, which is the *lower range* of global fallout in Colorado; thus the open space and residential areas located northwest and southwest of the 903 Pad were not significantly affected by RFP activity. Because of the limited number of measurements and the uncertainty associated with a unique value for global fallout in Colorado, the sequential Gaussian simulations modeling effort focused on the area east of RFP (east of State Plane Coordinate Projection 2093000).

A Gaussian model best fit the experimental variogram (Fig. 4) derived from the ccdf depicted in Fig. 3 and transformed into their normal score values. The Gaussian model represents a phenomenon with strong spatial continuity, typical of the wind dispersion that carried the plutonium particles across the soilscape.

The E-type estimates of the plutonium isotopic ratio, which are the point-by-point averages of the 100 realizations, are illustrated in Fig. 5. The E-type estimates of the plutonium isotopic ratio show a general west-east trend, characterized by a low isotopic ratio adjacent to the east perimeter of the plant, followed by a gradual increase in isotopic ratio estimates to the east, northeast, and southeast. The west-east pattern illustrated by the E-type estimates reflects wind dispersion consistent with the prevailing winds at RFP.

Litaor et al. (1995) constructed maps that showed the probability of exceedance of the mean global-fallout plutonium. Areas only 7 km east of RFP exhibited less than 20% probability of exceedance of the mean global fallout of 1.48 Bq kg^{-1} plutonium. However, as Krey (1976) clearly stated, some soil samples with greater plutonium activity may have little or no contribution from Rocky Flats, whereas samples with smaller plutonium activity than global fallout may contain some

Table 1. $^{240+239}Pu$ activity and the $^{240}Pu{:}^{239}Pu$ atomic ratio in individual samples near RFP (modified after Efurd et al. 1995).

ID[a]	$^{240+239}Pu \pm SD$ Bq kg^{-1}	$^{240}Pu{:}^{239}Pu \pm SD$
125	0.96 ± 0.015	0.16 ± 0.002
126	1.26 ± 0.022	0.14 ± 0.003
127	1.33 ± 0.033	0.128 ± 0.01
128	1.14 ± 0.019	0.143 ± 0.002
129	0.70 ± 0.019	0.16 ± 0.014
130	2.04 ± 0.030	0.163 ± 0.001
131	2.07 ± 0.033	0.114 ± 0.002
132	0.22 ± 0.004	0.139 ± 0.004
133	1.44 ± 0.033	0.162 ± 0.013
134	1.04 ± 0.044	0.101 ± 0.021
137	0.69 ± 0.111	0.155 ± 0.003
142	12.58 ± 0.222	0.055 ± 0.001
144	0.63 ± 0.007	0.107 ± 0.006
145	0.96 ± 0.370	0.094 ± 0.002
146	0.59 ± 0.100	0.115 ± 0.005
147	0.52 ± 0.011	0.165 ± 0.008
148	0.48 ± 0.011	0.097 ± 0.008
148	0.26 ± 0.007	0.136 ± 0.013
149	3.40 ± 0.056	0.112 ± 0.002
150	8.88 ± 0.148	0.064 ± 0.001
152	14.80 ± 0.370	0.065 ± 0.007
155	1.11 ± 0.037	0.108 ± 0.021
156	1.11 ± 0.019	0.13 ± 0.001
157	0.33 ± 0.007	0.116 ± 0.006
161	1.00 ± 0.019	0.075 ± 0.002
161	0.89 ± 0.019	0.069 ± 0.003
164	0.26 ± 0.007	0.136 ± 0.013
165	1.30 ± 0.026	0.112 ± 0.004
166	1.41 ± 0.037	0.106 ± 0.012
171	0.37 ± 0.015	0.089 ± 0.016
172	1.48 ± 0.037	0.083 ± 0.012
179	1.26 ± 0.037	0.099 ± 0.019
188	0.37 ± 0.007	0.108 ± 0.004
189	0.36 ± 0.011	0.097 ± 0.012
191	7.03 ± 0.137	0.069 ± 0.002
192	5.92 ± 0.074	0.06 ± 0.001
193	0.33 ± 0.007	0.125 ± 0.01
194	0.52 ± 0.011	0.087 ± 0.005
194	0.41 ± 0.022	0.079 ± 0.025
195	1.55 ± 0.026	0.113 ± 0.001
195	1.48 ± 0.026	0.115 ± 0.003
196	0.53 ± 0.011	0.117 ± 0.006

[a] See Fig. 1 for locations.

Rocky Flats plutonium. Hence, the present study was launched with the idea that the isotopic ratio data coupled with rigorous geostatistical analysis will provide a probability map with minimal ambiguity regarding the area impacted by RFP. Indeed, the E-type estimates depicted in Fig. 5 suggest that the area south of the plant exhibited isotopic ratios typical of global fallout plutonium and should be considered unaffected by RFP activity. On the other hand, most of the study area east of RFP exhibited values smaller than global-fallout plutonium (Fig. 5).

To evaluate this point further, a map that showed the probability of exceedance of the selected global-fallout isotopic ratio (i.e., 0.155) was computed (Fig. 6). This map showed that the probability of exceedance of a threshold of 0.155 east of RFP is extremely small, which means that most of the open space and residential areas in the study area were directly impacted by RFP activity.

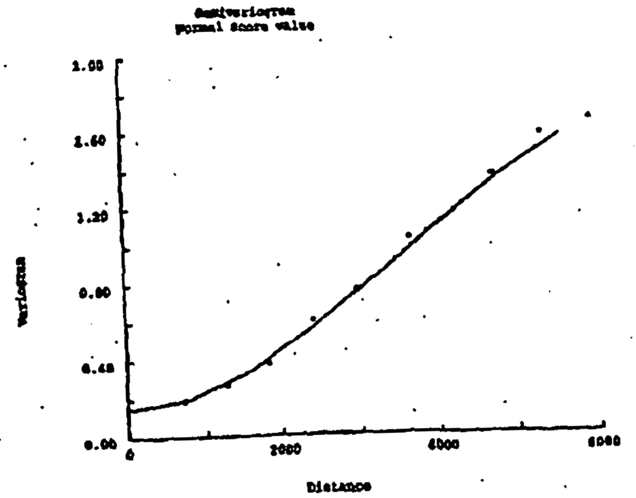

Fig. 4. The experimental variogram and fitted Gaussian model of the normal scores values. The variogram parameters are as follows: distance (range) = 5,000 m, sill = 2.0, nugget = 0.15, number of lags = 12, and unit separation distance between lags = 600 m.

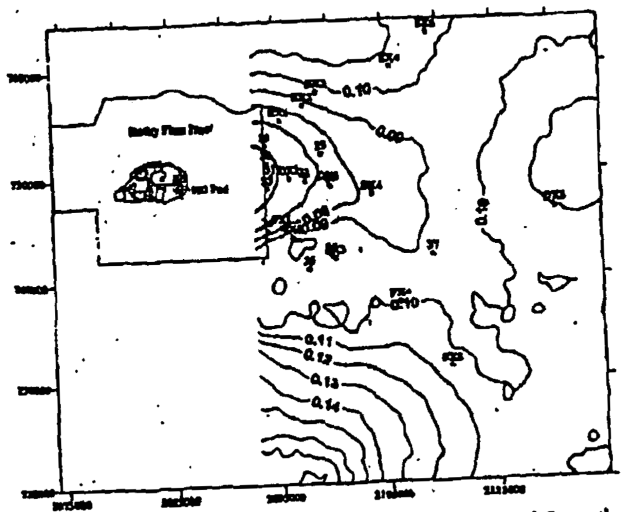

Fig. 5. E-type estimates of 100 Pu[(r)] realizations derived from the plutonium isotopic ratios.

The failure to delineate a definitive boundary to the areas impacted by RFP activity in the direction of the Greater Denver area did not result from the research design as outlined above but from unexpected small ccdf (see Figs. 2 and 3). The original research design called for the analysis of 63 sites across the study area for their plutonium isotopic ratio. Unfortunately, only 39 samples with unique coordinates were analyzed by Efurd et al. (1995) due to significant budget reduction across the DOE weapon complex. Critical locations in the southeast direction were not analyzed (see Fig. 2). Hence, the full

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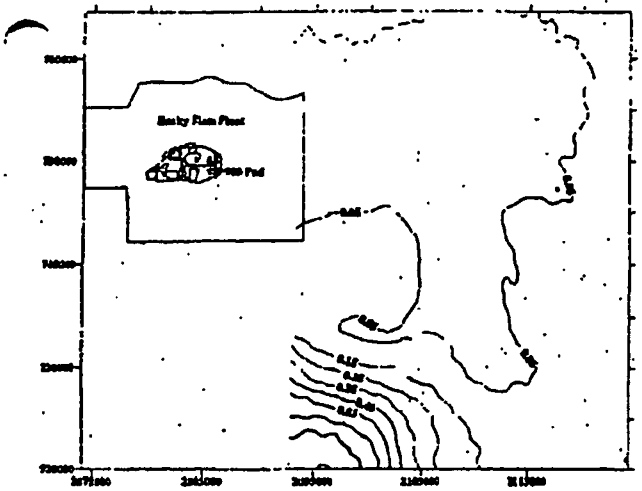

Fig. 6. A contour map of the probability that a given plutonium isotopic ratio exceeds the global fallout (i.e., 0.155) in Colorado.

extent of the area impacted by RFP is still undetermined. Furthermore, in a recent study, Ibrahim et al. (1997) found that soil samples collected 19 km directly east of RFP exhibited a plutonium isotopic ratio of 0.123 and plutonium activity of 1.1 Bq kg^{-1}. This finding indicates that RFP-derived plutonium has reached areas further to the east of the current study extent but with plutonium activity of less than the mean plutonium global-fallout.

Vertical distribution

The vertical distribution of plutonium activity in the 11 soil pits collected outside RFP is depicted in Fig. 7 and summarized in Table 2. The top layer (0–3 cm) is the most contaminated layer, with over 96% of $^{239+240}$Pu activity accounted for in the top 12 cm of the soil, whereas below the 12 cm depth plutonium activity decreased to background level. This distribution clearly attests to the general observed pattern of plutonium in the soil environment around RFP (e.g., Little et al. 1980; Litaor et al. 1994), which suggests that little downward movement of plutonium has occurred within these soils during the last 25 y. Several soils in the study area exhibited highly stony and coarse texture (not shown), which provides high potential for downward leaching. However, because $^{239+240}$Pu is strongly associated with organic matter and Fe oxides and hydroxides of the topsoil (Litaor and Ibrahim 1996), there has been little movement of plutonium down the soil profile regardless of soil type.

Plutonium loading analysis

The plutonium loading analysis on open space and residential areas was performed in three steps. In the first step, the planar area between isopleths of the E-type estimates (Fig. 5) was computed (Table 3). The area th-southeast of RFP that exhibited plutonium isotopic ratio greater than 0.155 with probability of exceedance of the mean plutonium global fallout ratio greater than 60%

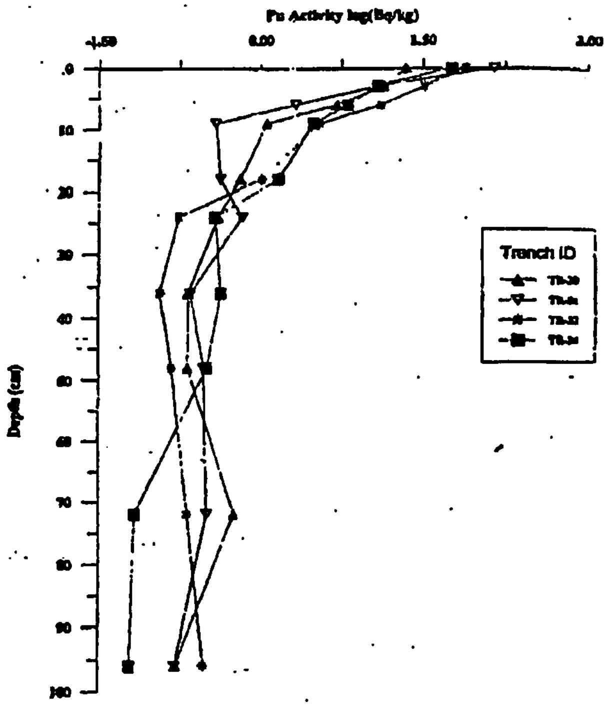

Fig. 7. Vertical profile of plutonium activity ($\log_{10}$ Bq kg^{-1}) in selected soil pits east of RFP.

(Fig. 6) was excluded from the planar area computations. In the second step, the plutonium activity at each soil depth (i.e., 0–3, 3–6, 6–9, and 9–12 cm) was averaged using the appropriate soil pits and macroplots bounded by a given isopleth interval and multiplied by the mean bulk density of the corresponding soil horizons times the planar areas computed above (Table 3). For example, the mean plutonium activity of Pits 30, 31, and 33 at the four soil depths times their mean bulk density times the planar area confined to the 0.06 isopleth yielded 2.76 GBq of plutonium. In the third step the mean plutonium activity in the topsoil layer (0–3 cm) in all soil pits and macroplots was adjusted by subtracting the mean plutonium global-fallout (1.48±0.5 Bq kg^{-1}) to yield the plutonium loading originated from RFP activity.

The mean total loading on open space and residential areas was 111.2 GBq with a standard error of estimate of 50.8 GBq (Table 3). The main source of uncertainty in the loading calculations across the study area resulted from high variability in plutonium activity over a small sampling scale. For example, in the area of 0.88 km^2 confined by the isopleth $\leq$ 0.06, the mean plutonium activity in the top sampling layer of the 3 representative pits was 80.3 Bq kg^{-1} with a standard deviation of 59.6 Bq kg^{-}, which resulted in a mean plutonium loading of 2.76 GBq and standard error of 1.11 GBq after adjusting to the estimated global fallout in Colorado. Similar variations in plutonium activity were observed across the study area in all depths of sampling

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Table 2. Plutonium activity and analytical error in 11 soil pits excavated east of RFP.

Location	TR-27	TR-28	TR-29	TR-30	TR-31	TR-32	TR-33	TR-34	TR-35	TR-36	TR-37
Depth (cm)						Bq kg^{-1}					
0–3	52.2 ± 8.6	20.2 ± 2.7	80.3 ± 10	22.2 ± 4.0	141 ± 22	106 ± 19	78.1 ± 10	58.9 ± 9	16.5 ± 4	35.2 ± 12	6.0 ± 1.4
3–6	9.8 ± 2.3	3.1 ± 1.1	7.5 ± 1.7	14.1 ± 3.6	12.2 ± 15	19.1 ± 3	32.9 ± 4	12.1 ± 3	24.2 ± 3	17.9 ± 5	1.8 ± 0.6
6–9	2.8 ± 1	0.6 ± 0.3	1.3 ± 0.6	5.0 ± 1.1	2.1 ± 0.7	6.3 ± 1.5	12.9 ± 2	6.4 ± 1.1	2.4 ± 0.8	12.6 ± 4	1.0 ± 0.5
9–12	0.6 ± 0.5	0.5 ± 0.3	0.6 ± 0.4	1.1 ± 0.6	0.4 ± 0.3	1.1 ± 0.4	3.5 ± 0.9	3.1 ± 1.4	11.4 ± 1.5	3.4 ± 1.2	0.5 ± 0.3
12–18	0.6 ± 0.7	0.3 ± 0.3	1.0 ± 0.2	0.6 ± 0.3	0.4 ± 0.3	0.3 ± 0.2	1.0 ± 0.5	1.5 ± 1.2	0.5 ± 2.5	4.0 ± 1.4	0.1 ± 0.3
18–24	0.5 ± 0.7	0.3 ± 0.2	0.4 ± 0.2	0.4 ± 0.5	0.7 ± 0.4	0.4 ± 0.2	0.2 ± 0.2	0.4 ± 0.7	0.3 ± 0.4	0.1 ± 0.4	0.2 ± 0.1
24–36	0.1 ± 0.5	0.1 ± 0.2	0.4 ± 0.3	0.2 ± 0.4	0.2 ± 0.2	0.5 ± 0.2	0.1 ± 0.2	0.4 ± 0.3	0.2 ± 0.2	0.6 ± 0.5	0.2 ± 0.2
36–48	0.1 ± 0.1	0.1 ± 0.2	0.6 ± 0.3	0.2 ± 0.3	0.3 ± 0.3	2.3 ± 0.7	0.1 ± 0.2	0.3 ± 0.4	0.1 ± 0.2	0.2 ± 0.3	2.6 ± 0.8
48–72	0.1 ± 0.1	0.1 ± 0.2	0.2 ± 0.2	0.6 ± 0.3	0.3 ± 0.2	0.9 ± 0.5	0.2 ± 0.1	0.1 ± 0.1	0.1 ± 0.2	0.1 ± 0.3	0.1 ± 0.1
72–96	0.6 ± 0.1	0.2 ± 0.3	0.6 ± 0.3	0.2 ± 0.1	0.2 ± 0.1	0.3 ± 0.2	0.3 ± 0.2	0.1 ± 0.3	0.1 ± 0.2	0.4 ± 0.4	0.1 ± 0.2

(Table 3). The large standard error of estimates (> 40%) in the inventory calculations must be taken into account for any dose reconstruction analysis or epidemiological study that employed plutonium distribution in soils (e.g., Johnson 1981). To demonstrate this point further, one can refer to a recent citizens' environmental sampling committee (CESC) report (1996), which was commissioned to produce a citizen-designed, off-site sampling

Table 3. Pu loading on open space and residential areas (see Fig. 4 for locations).

Depth cm	Pu activity Bq kg^{-1}	Bulk density g cm^{-3}	Planar area km^2	Pu loading GBq
Isotopic ratio ≤ 0.06; Pits 30, 31, 33				
	X SD			X SE*
0–3	80.28 ± 59.5	1.33 ± 0.17	0.88	2.76 ± 1.11
3–6	19.6 ± 11.1	1.33 ± 0.17	0.88	0.68 ± 0.18
6–9	6.6 ± 5.5	1.33 ± 0.17	0.88	0.23 ± 0.11
9–12	1.48 ± 1.48	1.33 ± 0.28	0.88	0.05 ± 0.29
Subtotal				3.72 ± 1.69
Isotopic ratio 0.06–0.07; Pits 28, 32 & Macroplot DX2				
0–3	52.88 ± 46.2	1.35 ± 0.23	2.99	6.22 ± 2.96
3–6	8.8 ± 8.5	1.35 ± 0.20	2.99	1.07 ± 0.37
6–9	2.2 ± 3.3	1.35 ± 0.20	2.99	0.26 ± 0.18
9–12	0.37 ± 0.37	1.35 ± 0.20	2.99	0.04 ± 0.02
Subtotal				7.59 ± 3.53
Isotopic ratio 0.07–0.08; Pits 27, 29, 34 & Macroplots DX3, EX1				
0–3	44.38 ± 29.6	1.59 ± 0.28	3.86	7.90 ± 2.59
3–6	6.6 ± 4.07	1.59 ± 0.25	3.86	1.22 ± 1.85
6–9	2.59 ± 1.85	1.59 ± 0.23	3.86	0.47 ± 1.11
9–12	1.11 ± 1.11	1.59 ± 0.23	3.86	0.20 ± 0.74
Subtotal				9.79 ± 6.29
Isotopic ratio 0.08–0.09; Macroplots DX4, EX2, FX1				
0–3	11.84 ± 5.18	1.24 ± 0.19	12.82	4.94 ± 1.48
3–6	1.48 ± 2.22	1.39 ± 0.15	12.82	0.79 ± 0.33
6–9	2.96 ± 4.44	1.39 ± 0.08	12.82	1.58 ± 1.48
9–12	0.74 ± 0.74	1.39 ± 0.08	12.82	0.39 ± 0.29
Subtotal				7.70 ± 3.58
Isotopic ratio 0.09–0.10; Pits 35, 36, 37 & Macroplots FX2, FX3, FX4 & EX3				
0–3	14.78 ± 12.58	1.5 ± 0.26	60.9	36.5 ± 11.8
3–6	8.88 ± 9.62	1.5 ± 0.19	60.9	24.3 ± 11.4
6–9	2.96 ± 4.44	1.5 ± 0.09	60.9	8.11 ± 4.8
9–12	2.59 ± 4.07	1.6 ± 0.17	60.9	7.57 ± 4.8
Subtotal				76.4 ± 32.8
Isotopic ratio ≥ 0.10; Macroplots EX4, EX5, DX5, FX5				
0–21	0.59 ± 0.48	1.36 ± 0.07	36.01	6.08 ± 2.96
Total	-		117.4	111.2 ± 50.8

* The standard error of estimate (SE) was computed using the method described by Gilbert (1987).

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Table 4. Classification analysis that tested the agreement between the measured Pu isotopic ratio (Ibrahim et al. 1997) and estimated ratios derived from the sGs (present study).

ID[a]	Measured	Estimated	Outcome
FX-5	0.106	0.10–0.11	True
FX-4	0.079	0.10	False
FX-3	0.066	0.09–0.10	False
DX-5	0.082	0.11	False
DX-3	0.06	0.07–0.08	False
EX-5	0.105	0.10–0.11	True
EX-4	0.089	0.10–0.11	False

[a] See Fig. 5 for location.

program near RFP, and the study by Ibrahim et al. (1997). The CESC sampled 24 sites within the area east and southeast of RFP and used the results to test the accuracy of several published plutonium maps. For example, the results of the 24 sites were superimposed on the E-type estimates map generated from the comprehensive data set of 852 soil samples (Litaor et al. 1995). Despite the comprehensive compilation of plutonium data and the rigorous geostatistical treatment that produced the map, only 19 of the 24 sites (79%) were classified correctly using the E-type map. Five sites exceeded the published isopleths by 0.7 to 125 Bq kg^{-1} indicating the spotty nature of plutonium activity in the soil environs across the small sampling scale.

Ibrahim et al. (1997) measured the plutonium isotopic ratios in several sites depicted in Fig. 5, and their results were compared with the estimated isopleths (Table 4). Only two locations (28%) were classified correctly using the E-type map generated from the simulations. This poor agreement probably resulted from the small ccdf used in the simulation and the vastly different soil sampling techniques employed. In the present study, each soil sample was collected over a 4.05-ha plot by sampling 25 subsamples evenly distributed over the entire plot to a depth of 0.64 cm (Litaor et al. 1995). The 25 subsamples were composited to represent the entire plot. The soil sampling by Ibrahim et al. (also see Webb et al. 1994, 1997) was done in microplots of 1 × 1 meter size, with depth intervals of 3 cm.

Notwithstanding the spatial uncertainty inherited in any study that attempted to calculate the loading of radionuclides on a large area; the present plutonium loading is fairly similar to that found in an earlier study by Krey (1976), who reported 125.8±33.3 GBq of plutonium deposited across a considerably larger area, which included the entire Denver area. The similar results of the plutonium loading analyses, which used vastly different methods in assessing the aerial extent and plutonium activity in the soil, suggest that most of the plutonium released by RFP resides in open space and residential areas that are in close proximity to the plant perimeter (i.e., within the boundaries of the present study). These findings also illuminated the scale issue that governs most radionuclide distribution in the soil environs of all contaminated sites. We may not be able to produce a single map that will always classify correctly the plutonium activity and/or the plutonium isotopic ratio, but we can provide an upper limit to the amount of plutonium released by RFP.

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